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SYNTHESIS AND STUDY OF MATERIALS FOR SUPERCONDUCTING
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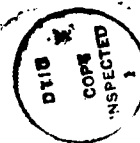


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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Study of high-T_c superconductors in single-crystal and thin-film form has begun. Investigation of semiconductor tunnel barriers has led to improvement of their theoretical description. Transport measurements of layered films of transition metal nitrides showed evidence of spin-orbit scattering and dimensional crossover effects. A technique for analyzing tunneling conductances to obtain the amount of Zeeman splitting using Fourier transforms has been developed and applied to V₃Ga tunneling data. Spin-orbit scattering rates measured by two independent techniques, spin-polarized tunneling and		

magnetoresistance, have been shown to be similar. The strength of Fermi-liquid corrections on the effective magnetic moment of conduction electrons in vanadium and gallium has been found using spin-polarized tunneling. The effect of spin-orbit scattering on the density of states of superconducting thin films has been measured. The superconducting properties of ultra-thin niobium films were studied.



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I. Research Objectives

AFOSR-TR. 88-0453

The objectives of the research under this grant were as follows:

- a. Synthesize high transition temperature superconducting materials potentially useful in superconducting electronics.
- b. Characterize the physical and chemical properties of the materials and their surfaces with ESCA and other analytical instrumentation.
- c. Form tunnel junctions on thin films of these materials and investigate their properties.
- d. Make spin-polarized and electron-phonon spectroscopy measurements on superconducting thin films.
- e. Evolve practical theories to explain the properties of the superconducting materials and tunnel junctions.
- f. Attempt to make epitaxial films of the superconductors on crystal substrates to improve their characteristics.
- g. Explore superconductor-insulator and superconductor-superconductor composite layered structures.
- h. Investigate ultra-thin superconducting films to observe surface and localization effects.
- i. Form artificial tunnel barriers of refractory insulators compatible with high temperature superconductors.
- j. Compare results with basic theory including Fermi liquid effects.
- k. Explore promising unusual materials.

II. Status of Research Effort

The research program has concentrated on fundamental properties of promising superconducting and tunnel barrier materials which may prove useful for superconducting electronics.

1. High T_c Materials

We have fabricated high-quality specimens of $\text{YBa}_2\text{Cu}_3\text{O}_7$ in ceramic, single-crystal and thin-film form. Measurements of transport properties were made in high magnetic fields to elucidate the nature of the superconducting and normal states of this material. Tunneling studies were begun using vacuum barrier techniques. We have found that thermodynamic fluctuations are important in determining the shape of the resistive transition and that the critical field depends strongly (\sim a factor of 4) on magnetic field direction with respect to the crystal axes. This work is continuing.

2. Extension of Simmons' Theory to Tunneling through Small Barriers and Its Application to Amorphous Ge

Using the WKB approximation Simmons developed an expression for the current as a function of applied voltage through a rectangular barrier [1]. In doing this he ignored several terms which become important in the limit of small barriers. We have simply retained these terms to obtain a more accurate analytic expression in that limit. Two expressions were

developed: one for $V < \phi$ and one for $V > \phi$, where ϕ is the barrier height and V the applied voltage. The improvement due to using the "extended theory" becomes more marked as the barrier height is lowered. In fact, Simmons' original expression gives negative values for the tunneling current for high enough voltages. The terms which Simmons ignored become important when $\bar{\phi} \leq eV$ or $\frac{\sqrt{2m\bar{\phi}}}{h} \leq 1/s$ where $\bar{\phi}$ is the mean height of the barrier over the negatively biased electrode and s is the distance between the classical turning points. a-Ge tunnel barriers are "small" by the above criteria. We found that their $I(V)$ curves could be better fit with the extension to the theory than without, although not perfectly.

[1] J.G. Simmons, J. Appl. Phys. 34, 1793 (1963).

3. Spin-Orbit Scattering Measurements from Localization and Superconducting Tunneling

Spin-orbit scattering affects electronic transport in metals in both the superconducting and normal state. In the superconducting state, spin-orbit scattering causes spin mixing between the time-reversed states that form the Cooper pairs. Superconducting measurements of the upper critical field and the tunneling density of states have yielded a value for the spin-orbit scattering rate by using the Fermi liquid theory of superconductivity [1]. In the normal state, spin-orbit scattering affects the random potential scattering that causes quantum interference among electrons. Normal state measurements of the magnetoresistance have yielded values for the spin-orbit scattering rate by using weak localization theory [2-4]. In the past, each of these two methods individually have given

internally consistent measurements of the spin-orbit scattering rate for a number of metals. In this experiment both superconducting and normal state measurements have been done on the same thin Al film sample, and, hence, we directly compare for the first time the rates of spin-orbit scattering. Our main conclusion is that the two methods give rates of spin-orbit scattering that agree reasonably well. This agreement is particularly surprising considering the diverse nature of the experiments, the different ranges of magnetic field and temperature used, and the absence of quantum transport corrections in the superconducting theory.

- [1] J.A.X. Alexander, T.P. Orlando, D. Rainer, and P.M. Tedrow, Phys. Rev. B31, 5811 (1985).
- [2] G. Bergmann, Phys. Rev. B29, 6114 (1984).
- [3] J.M. Gordon, C.J. Lobb, and M. Tinkham, Phys. Rev. B28, 4046 (1983).
- [4] P. Santhanam and D.E. Prober, Phys. Rev. B29, 3733 (1984).

4. Properties of Ultra Thin Nb Films

Niobium is the highest T_c elemental superconductor, and Nb films are very resilient, making them an interesting candidate for electronic applications. In the past the T_c 's of Nb films have been found to be greatly suppressed due to native suboxide formation. In addition, these suboxides have made junction fabrication very difficult.

We have made Nb films from 30-150 Å thick with T_c 's from 3.5 to 9.0 K using the oxidized metal overlayer method to suppress native oxide formation [1]. The Nb was electron beam evaporated onto 700°C sapphire

substrates followed by deposition of 25 Å of aluminum at 220°C. We find T_c has a (thickness)⁻¹ dependence which is consistent with both proximity effects and weak localization effects. We found our films' resistivity and resistance per square could vary more than 50% for a given thickness with no effect on T_c or H_c , telling us that the superconducting properties are determined within the grains while transport properties are intergrain effects.

Critical field data were taken on these films. $H_{c||}$ follows a \sqrt{T} law near T_c as is expected. The perpendicular critical fields for the same films, however, show a marked curvature near T_c which has also been seen in other 2-d systems [2]. These results have not been explained. However, $H_{c\perp}$ ($T=0$) goes as $1/d$ from 50 Å to 150 Å as one would expect from Ginsberg-Landau theory, assuming the coherence length ξ is independent of film thickness (and hence T_c). Likewise, $H_{c||}$ ($T=0$) $\sim \frac{1}{d^{3/2}}$, again consistent with Ginsberg-Landau theory if we assume ξ is independent of d . Below 50 Å $H_{c||}$ and $H_{c\perp}$ become constant, independent of film thickness at 14 tesla and 8 tesla, respectively. Weak localization effects appear to play an important role in these films.

In conclusion, disorder and weak localization have important effects on both T_c and critical fields which need to be understood to use ultra thin films in electronics applications.

[1] M. Gurvitch and J. Kow, *Advances in Cryogenic Engineering* 30, (1984).

[2] J. Graybeal and M.R. Beasley, *Phys. Rev.* B29, 4167 (1984).

5. The Effect of Spin-Orbit Scattering on the

Spinstates of Pauli-limited Superconductors

Aluminum films about 4 nm thick are very convenient for studying the effects of spin-orbit scattering on high field superconductivity. A pure Al film has a small spin-orbit scattering rate, and overcoating Al with Pt has been found to cause an increase in λ_{so} [1]. Thus, a wide range of λ_{so} can be investigated. In addition, tunnel junctions are formed more easily on Al films than on any other superconductor. Previous studies [1] of Al(Pt) films showed that the parallel critical magnetic field and total tunneling conductance behaved in a way consistent with theoretical expectations if it was assumed that increasing Pt coverage simply increased λ_{so} . We have now applied the technique of spin-resolved spin-polarized tunneling [2] to examine in detail the spin states of the quasiparticles in the superconducting state for various values of λ_{so} in high magnetic fields. This technique involves measuring the conductance of an Fe-superconductor tunnel junction in an applied field. The individual spinstates of the superconductor can then be obtained by simple algebraic manipulation.

In the absence of spin-orbit scattering, the individual conductances from spin-up and spin-down electrons in a magnetic field are identical in shape but shifted with respect to each other by the Zeeman energy δ , which near the phase boundary is $g\mu_B H_{ext}(1-G^0)^{-1}$ [3]. Here g is the g -factor of the electron in the metal, H_{ext} is the applied field and G^0 is the antisymmetric Fermi liquid parameter. We have previously shown that for Al $g = 2.0$ and $G^0 = 0.3$ [4]. The reason that the individual conductances are identical in shape is that in the absence of spin-orbit scattering the superconductivity pairing between electrons which are in an eigenstate of spin, i.e., the Cooper pairing is between spin-up and spin-down elec-

trons. In zero field the conductance due to each spin state is equal. In a field the electrons just shift their energy by the Zeeman splitting so that the conductance at a given energy is due to both spin-up and spin-down electrons that are displaced by just the Zeeman energy difference.

In the presence of spin-orbit scattering, the superconducting pairing is no longer between electronic states which are eigenstates of spin, i.e., the Cooper pairing is between time-reversed states with a mixing of spin-up and spin-down electrons. The mixing is apparent for a thin film of Al which has a large coverage of Pt and hence a large spin-orbit scattering rate ($\lambda_{so} > 3$). The thin-film was measured in 4 tesla at 0.6 K. The shapes of the individual spin conductances are no longer identical and shifted, showing clearly that the conductance at a given energy is due to both spin-up and spin-down electrons that are not displaced by just the Zeeman splitting.

- [1] P.M. Tedrow and R. Meservey, Phys. Rev. B25, 171 (1982).
- [2] P.M. Tedrow, J.S. Moodera, and R. Meservey, Solid State Commun. 44, 587 (1984).
- [3] J.A.X. Alexander, P.M. Tedrow, D. Rainer, and T.P. Orlando, Phys. Rev. B, 1985.
- [4] P.M. Tedrow, J.T. Kucera, D. Rainer, and T.P. Orlando, Phys. Rev. Lett. 52, 1637 (1984).

6. Investigation of Fermi-liquid Effects in Superconducting Vanadium and Amorphous Gallium by Spin-polarized Tunneling

Fermi-liquid renormalization effects in are difficult to measure in most normal metals. However, it has been shown [1] that the parameters which characterize them can be determined in superconductors by using spin-polarized tunneling.

When evaporated onto a substrate held at liquid helium temperature, gallium forms an amorphous phase with a large electron-phonon coupling constant [2] ($\lambda_{ep} \sim 2.2$). This should make the many-body effects large and easily observable. Furthermore, gallium is a relatively low atomic number element and should have a long spin-orbit scattering time. The Zeeman splitting of the quasi-particle states in amorphous gallium has previously been observed in Al/Al₂O₃/a-Ga junctions by Tedrow and Meservey [3]. We are attempting to measure accurately this splitting using the technique of spin-polarized tunneling. The Zeeman splitting is renormalized by a factor of $(1+G_0)^{-1}$ near a second-order phase transition to the normal state. This allows a measurement of the $l=0$ anti-symmetric Fermi-liquid parameter, G_0 .

We have successfully carried out measurements on Al/Al₂O₃/Ga junctions and are evaluating the conductance results to obtain G_0 . We would like to compare the Fermi-liquid effects in a-Ga with those already observed in vanadium. Vanadium is of interest because it is a possible spin-fluctuation system so that there may be an electron-electron as well as an electron-phonon contribution to the Fermi-liquid interaction. Preliminary spin-polarized tunneling measurements on vanadium indicate a value for G_0 on the order of 0.2. This is within the range of values consistent with our critical field data. It is also consistent with previously determined values for λ_{ep} , λ_{ee} (electron-electron coupling constant) and \bar{I} (the Stoner factor).

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- [2] T.T. Chen, J.T. Chen, J.D. Leslie, and H.J.T. Smith, Phys. Rev. Lett. 22, 526 (1969).
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7. Determination of the Zeeman Splitting
in Superconductors by Fourier Analysis of Tunneling Data

The measurement of G^0 is particularly interesting in the high- T_c vanadium based compounds, because the critical field behavior cannot be understood in the framework of the conventional theory without including fermi-liquid effects. As an example, we applied this analysis to the technologically important A15 compound V_3Ga and showed that the g-factor is approximately 2.0.

One method to directly measure the Zeeman splitting in spite of the thermal smearing and field broadening is to use the technique of spin-sensitive tunneling [1] in which the electrons tunnel into the superconductor from a ferromagnetic counterelectrode. The spin-sensitive tunneling technique, however, has not been widely used on other materials up to now because it is very difficult to make reliable tunnel junctions using a ferromagnet as a counterelectrode.

Our method to directly measure the Zeeman splitting, in spite of the thermal smearing and field broadening, is a technique of Fourier analysis, known as cepstral analysis, of the conductance of tunnel junctions with a normal counterelectrode. This method can be used to determine the Zeeman splitting even in cases where the conductance is considerably washed out.

The advantage of this method is that it does not require a ferromagnetic counterelectrode.

This method has been used to determine the Zeeman splitting in V_3Ga thin films.

The films contain 23%, 25% and 28% gallium, respectively. As a consequence of the difference in composition, the superconducting critical temperature T_c and the energy gap also vary, with the stoichiometric composition with 25% gallium having the highest energy gap. It is, however, very difficult to examine the value for the Zeeman splitting by looking at the measured conductances because they just barely intimate a separation into two peaks; however, in the cepstra for these same three sets of tunneling data, the peaks corresponding to the Zeeman splitting can be clearly seen. The Zeeman splitting is near the expected value of 1.2 meV for all compositions and seems to be smallest for the stoichiometric composition. Here the g-factor for V_3Ga is 2.0 approximately.

The success of the cepstral analysis in determining the Zeeman splitting rests on the fact that such analysis gives a separate signature due to the Zeeman splitting, the density of states and the thermal smearing.

It should be noted that these methods are possible because efficient algorithms exist for implementing Fourier analysis which demand neither long computer time nor much computer memory. All our calculations were done on an HP87 personal computer.

- [1] P.M. Tedrow, J.S. Moodera, and R. Meservey, Solid State Commun. 44, 487 (1982).
- [2] S.J. Bending, M.R. Beasley, and C.C. Tsuei, Phys. Rev. B30, 6342 (1984).

8. Study of Superconducting Layered and Coevaporated (V/Mo)N Films

Layered films of V/Mo with layer thicknesses from 10 to 400 Å (overall thickness ~2500 Å) were made by e-beam evaporation and subsequently nitrided in situ at ~700°C. The parallel and perpendicular critical fields were measured as a function of temperature and compared with the theory of Takahashi and Tachiki [1]. Both 3D+2D and 2D+2D dimensional crossover effects were observed. The 2D+2D temperature dependence crossover is a new effect predicted by Takahashi and Tachiki for multilayers in which the constituent layers have a large ratio of diffusion constants.

Coevaporated V(Mo)N films with various Mo concentrations were also made. Their critical fields were compared with the layered (V/Mo)N films. There is some indication that the spin-orbit scattering rate is higher in the layered films than in those that were coevaporated. This suggests that high-z impurities are more effective spin-orbit scatterers at surfaces than in the bulk.

The effect of Mo concentration on the T_c of V(Mo)N was also studied. In no case was a T_c greater than that of pure VN obtained. This is of interest because of the close lattice match of VN with the postulated high T_c phase of MoN.

- [1] S. Takahashi and M. Tachiki, Phys. Rev. B 33, 4620 (1986); *ibid*, 34, 3162 (1986).

3. List of Publications

P.M. Tedrow and R. Meservey

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LT17, U. Eckern, A. Schmid, W. Weber, and H. Wuhl, Eds. (Elsevier Science
Publishers B.V., 1984), p. 837

Vanadium Nitride Spin-Orbit Scattering Rate Measured Using Spin-Polarized
Tunneling.

P.M. Tedrow, J.T. Kucera, D. Rainer, and T.P. Orlando

Phys. Rev. Lett. 52, 1637 (1984)

Spin Polarized Tunneling Measurement of the Anti-symmetric Fermi-Liquid
Parameter G^o and Renormalization of the Pauli Limiting Field in Al.

G.A. Gibson and R. Meservey

J. Appl. Phys. 58, 1584 (1985)

Properties of Amorphous Germanium Tunnel Barriers

J.S. Moodera, R. Meservey, and P.M. Tedrow

IEEE Trans. Magn. MAG-21, 551 (1985)

Determination of Thin Penetration Depth of Type II Superconducting Films

G.B. Hertel and T.P. Orlando

Phys. Rev. B 32, 166 (1985)

Determination of the Zeeman Splitting in Superconductors by Fourier Analysis
of Tunneling Data

J.A.X. Alexander, D. Rainer, and P.M. Tedrow

Phys. Rev. B 31, 5811 (1985)

Theory of Fermi-Liquid Effects in High-Field Tunneling

P.M. Tedrow, J.E. Tkaczyk, and A. Kumar

Phys. Rev. Lett. 56, 1746 (1986)

Spin Polarized Electron Tunneling Study of An Artificially Layered Super-
conductor with Internal Magnetic Field: EuO-Al.

J.H. Quateman

Phys. Rev. B 34, 148 (1986)

T_c Suppression and Critical Fields in Superconducting Nb Films.

G.A. Gibson and R. Meservey

Bull. Am. Phys. Soc. 31, 437 (1986)

Spin-Orbit and Fermi Liquid Effects in Superconducting Vanadium Films.

J.S. Moodera and R. Meservey

Bull. Am. Phys. Soc. 31, 239 (1986)

Superconductivity in Bi and Ga Films Deposited at 300 K or 77 K Over Very Thin Ni Layers.

J.S. Moodera, P.M. Tedrow, and R. Meservey

Advances in Cryogenic Engineering Materials, Vol. 32

Edited by R.P. Reed and A.F. Clark, (1986), p. 679.

Critical Field Studies of Reactively Sputtered and Nitrided NbN, VN, and V(Mo)N Films

J.A.X. Alexander, P.M. Tedrow, and T.P. Orlando

Phys. Rev. B34, 8157 (1986)

Spin-orbit Scattering Measurements from Localization and Superconducting Tunneling

J.S. Moodera, P.M. Tedrow, and J.E. Tkaczyk

Phys. Rev. B36, 8329 (1987)

High Magnetic Field Study of Superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

J.S. Moodera, R. Meservey, J.E. Tkaczyk, C.X. Hao, G.A. Gibson, and P.M. Tedrow

Phys. Rev. B (to be published)

Critical Magnetic Field Anisotropy in Single Crystal $\text{YBa}_2\text{Cu}_3\text{O}_7$

G. Gibson, J.S. Moodera, R. Meservey, and P.M. Tedrow

Phys. Rev. B (to be submitted)

Effect of Growth Morphology, Layering and Dimensional Cross-over on H_{c2} of Transition Metal Nitrides

G. Gibson, R. Meservey, and P.M. Tedrow

Phys. Rev. B (to be submitted)

Fermi-liquid Renormalization of H_{c2} of Vanadium and Gallium Thin Films

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